Pressure Effects and Large Polarons in Layered MgB_2 Superconductor

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We consider the dependence of the MgB_2 superconducting critical temperature on the pressure. Our model exploits the influence of the large polarons on the band structure of the layered MgB_2 superconductor. Namely, the hole Pekar-Fröhlich polarons form quasi two-dimensional potential wells in the boron plane which shift the positions of the σ- and π-bands. This energy shift depends on the pressure and the Cooper pairing of the correlated σ-electrons happens inside polaron wells. The results obtained are as follows: $dT_c/dp \simeq -\alpha(5.2 \pm 0.9)\ K/GPa$ or $dT_c/dp \simeq -\alpha(6.9 \pm 1.1)\ K/GPa$ for a different choice of the Grüneisen parameter. Being compared with known experimental data they give us a resonable interval for the value of the Fröhlich electron-phonon coupling constant: $\alpha \simeq 0.15 - 0.45$.

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I. INTRODUCTION

Soon after the discovery of the superconductivity in MgB_2 the pressure influence on the critical temperature T_c has been experimentally studied. The baric derivatives dT_c/dp were extracted from the obtained data. At the low-pressure ($p \leq 2\,GPa$) the experimental results are as follows: $dT_c/dp = -1.2\ K/GPa$ in Refs. 1,2 and $-2\ K/GPa$ in Refs. 1,3,4. Experiments at high-pressure lead at the same sign of the baric derivatives but demonstrate lower values. Namely, $dT_c/dp = (-0.8, -1.07, -1.11) K/GPa$ in Refs. 5,1,6, respectively. Many conventional low-T_c superconductors also reveal the same sign, $dT_c/dp < 0$, but their absolute values are much lower.

Borons in the crystal MgB_2 are packed in honeycomb layers alternating with hexagonal layers of magnesium ions. The in-plane distance, $b = 1.78$ Å, between the boron cations is less than those between magnesium anions, $a = 3.08$ Å. The space separating the boron planes is of the size $c = 3.52$ Å. The magnesium ions are positioned above the centers of hexagons formed by boron sites and donate their electrons to the boron planes: $Mg^{2+}\left[B^{-}\left(p^2\right)\right]_2$. These p-electrons form σ- and π-bands and the charged Mg-layers lift the three-fold degeneracy between σ- and π-electrons and shift the electronic energy bands so that π-band becomes lower than the σ-band. It occurs that the latter crosses the Fermi-level providing the light and heavy hole formation. The electronic structure is formed by the narrow energy-band of the two-fold degenerate σ-electrons and the wide-band of the π-electrons.

As to the phonon modes in MgB_2 they have a sharp cut-off at about 100 meV. Below this energy it is supposed the existence of different phonon modes[7]. Note that the phonon energies mentioned in Ref. 7 could be overestimated because the approximation used to derive them is valid only for cubic lattices[8,9]. The anharmonic in-plane breathing mode E_{2g} (74.5 meV) strongly interacts with the narrow band σ-electrons[10]. Due to this fact its energy is lower than that of the out-of-plane tilting boron mode B_{1g} (87.1 meV). Besides, there are low-energy acoustic modes and indications are also found[11–13] of the existence of phonon modes with extremely low energies.

The most of the theoretical speculations on the T_c pressure dependence in MgB_2 are based on the semi-phenomenological approaches and do not involve the microscopic arguments. In the scope of the conventional BCS theory dT_c/dp is governed by the competition between a decrease of the density of electronic states at the Fermi level and an increase of the Debye frequency in the BCS formula for T_c. The weak electron-phonon coupling constant[14] in MgB_2 and the absence of the peak in the electronic density of states at the vicinity of the Fermi-energy[15] cannot provide the BCS scenario.

On the other hand, the spectroscopic measurements of the superconducting gap[16–18], the specific heat behavior[19,20], the low isotope effect[21,22] (under the substitution of both B and Mg isotopes) and the pressure effects[23] evidence in favor of the complex mechanism of the superconductivity in MgB_2. The calculated spectral function[24] and the analysis[14] of the reflectance measurements[25] show the possibilities of different superconducting mechanisms beyond the conventional electron-phonon BCS pairing.

The positive baric derivative of T_c was found[26,27] in the microscopic model of the hole dressed superconductivity what opposes the experimental data.

Among optical modes in MgB_2 the E_{2g} is the only Raman active mode. The high-pressure (up to 15 GPa) Raman experiment[28] has revealed a large pressure shift of the E_{2g} phonon mode. The E_{2g} mode does not manifest any softening at T_c (see Ref. 29) therefore does not provide the superconducting electron pairing. Neverthe-

less, it is responsible for the T_c behavior with pressure as we will see later. It should be pointed out that baric experiments have been completed both for pressed powder and single crystals. No structural transitions were found in experiments[3,28,30] up to the pressure 40 GPa.

The goal of the present paper is to calculate the baric derivative dT_c/dp. We suppose that the large polarons are formed in the boron-planes due to the σ-electrons interaction with the optical E_{2g}-phonons[7]. Because of the polaron formation the σ-band becomes lower while the π-electrons are not influenced. As the events take place in the layered medium we consider the quasi-two-dimensional polaron. The polaron anisotropy parameter and, correspondingly, the σ-band energy shift depend on the lattice constants influenced by the pressure. Roughly speaking, the Cooper pairs of the correlated σ-electrons in the MgB_2 superconducting condensate are plunged into the pancake polaron. To treat the Cooper paring we accept the kinematic mechanism[31,32] in MgB_2.

II. FORMULATION OF THE MODEL

In order to describe the superconducting state of MgB_2, we adopt the model with the strongly interacting σ-electrons (with a narrow bandwidth $2w_1$) overlapping with the noncorrelated π-electrons (with a wide bandwidth $2w_2$) of boron ions. The important parameter here is the distance r between the centers of σ- and π-bands influenced by the pressure.

The positive Hall current and the thermoelectric power measurements[33] indicate the hole conductivity in MgB_2. The polaron radius is given by the expression

$$R \sim \sqrt{\frac{\hbar}{m\omega}} = \frac{87.3\ \mathring{A}}{\sqrt{(m/m_e)\cdot(\hbar\omega/1\ meV)}}, \qquad (1)$$

where m and m_e are the hole and the vacuum electron masses, respectively, and $\hbar\omega$ is the low-frequency optical phonon energy responsible for the polaron formation. Inserting the value $\hbar\omega \simeq 74.5\ meV$ for the E_{2g}-phonons and the light hole band mass[34] $m = 0.25m_e$ into Eq. (1) the polaron radius can be estimated as $R \simeq 20\ \mathring{A}$ which is much larger than the lattice constant.

Studying the polaron sector of the system we apply the conventional Pekar-Fröhlich Hamiltonian generalized for the consideration of the peculiarities of MgB_2. To take into account the fact that the σ-electrons are located in narrow layers inhabited by boron atoms we introduce the in-plane mass m which is the light hole band mass and the large mass M in the orthogonal (z) direction. The ratio m/M of these masses is related to the probability of a carrier to tunnel from one layer to another.

To take into account the different polarizability of the medium in the xy-plane and in the z-direction we suggest to simulate it by using the anisotropic Columb potential for the interaction of the hole with the induced polarization field. That is, we use the potential of the form $V(\vec{r}) \propto 1/\sqrt{x^2+y^2+(z/\gamma)^2}$ with $\gamma \leq 1$ being a phenomenological parameter to describe the deformation of the Coulomb potential along the z-axis (that is, the oblation of the equipotential surface). In the momentum space the potential is of the form $\tilde{V}(\vec{k}) \propto 1/(k_\parallel^2 + \gamma^2 k_z^2)$.

In a sense we study the electronic sector as how all events happen to be located inside the large polarons. The large polaron being formed we obtain the lowering of the energy of carriers, that is the renormalization of the parameter r which becomes dependent on the applied pressure. This influences drastically the electronic structure of MgB_2-like systems, especially the relative position of σ- and π-bands.

A. Polaronic Sector

Thus, we consider an anisotropic polaron whose motion is confined to the xy-plane while the motion in the perpendicular z-direction (along the crystallographic c-axis of MgB_2) is restricted. The Pekar-Fröhlich Hamiltonian reads as follows:

$$H = \frac{\vec{p}_\parallel^{\,2}}{2m} + \frac{p_z^2}{2M} + \hbar\omega \sum_{\vec{k}} a_{\vec{k}}^\dagger a_{\vec{k}} + \frac{1}{\sqrt{\Omega}} \sum_{\vec{k}} \left(a_{\vec{k}}\, V_{\vec{k}}\, e^{i\vec{k}\cdot\vec{r}} + a_{\vec{k}}^\dagger\, V_{\vec{k}}^*\, e^{-i\vec{k}\cdot\vec{r}} \right), \qquad (2)$$

where Ω is the crystal volume, $\vec{p}_\parallel, p_z$ are the hole momenta in the xy-plane and in the orthogonal z-direction, respectively, and ω is the LO-phonons frequency. Operators $a_{\vec{k}}^\dagger(a_{\vec{k}})$ are the creation (annihilation) operators of the phonons with a momentum $\vec{k}$. The electron-phonon interaction term is specified by $V_{\vec{k}}$; its squared modulus is the Fourier transform of the Coulomb potential $V(\vec{r})$. In contrast with the conventional definition we introduce an additional anisotropy of the Coulomb potential to describe the different polarizability of the medium in various directions (in the isotropic boron plane and perpendicular to it):

$$V_{\vec{k}} = -i\hbar\omega \left(\frac{4\pi\alpha}{k_\parallel^2 + \gamma^2 k_z^2} \sqrt{\frac{\hbar}{2m\omega}} \right)^{1/2}. \qquad (3)$$

Here α is the conventional Fröhlich coupling constant of the electron-phonon interaction.

The MgB_2 baric compression is anisotropic[2,28,35]. According to Ref. 28 the compressibility along c-axis almost twice larger the plane compressibility. Under the hydrostatic pressure the initial compression along the c axis is larger than along the boron plane[36]. We estimate the parameter γ as the ratio of the standard lattice constants: $\gamma \sim c/2a$ (here $c/2$ is the distance between charged boron and magnesium planes). When the pressure increases the distance between the Mg and the B

planes decreases which is described by the decreasing of the Coulomb potential anisotropy γ in our model. The polaron self-energy ΔE can be found within the second order of the perturbation theory:

$$\Delta E = -\frac{\alpha\hbar\omega}{2\pi^2}\int \frac{d^3k}{(k_\parallel^2+\gamma^2k_z^2)(k_\parallel^2+\frac{m}{M}k_z^2+1)}. \qquad (4)$$

The integration in Eq. (4) being performed we arrive at the expression:

$$\Delta E = -\alpha\hbar\omega\sqrt{\frac{M}{m}}\begin{cases} \frac{1}{\sqrt{1-\Gamma^2}}\ln\frac{1+\sqrt{1-\Gamma^2}}{\Gamma}, & \Gamma\le 1; \\ \frac{1}{\sqrt{\Gamma^2-1}}\arctan\sqrt{\Gamma^2-1}, & \Gamma\ge 1. \end{cases} \qquad (5)$$

Here the parameter $\Gamma = \gamma\sqrt{M/m}$. This formula reproduces two well-known limiting cases. When $M = m$ and $\gamma = 1$, then $\Gamma = 1$ and we obtain $\Delta E = -\alpha\hbar\omega$, that is the conventional result for the three-dimensional polaron. When $\gamma = 1$ and $M \gg m$, we arrive at the result $\Delta E = -\frac{\pi}{2}\alpha\hbar\omega$ which is valid for the two-dimensional polaron confined to a plane[37,38]. If γ is finite and $M \gg m$, then $\Gamma \gg 1$ and Eq. (5) leads at the following expression:

$$\Delta E = -\frac{\pi}{2\gamma}\alpha\hbar\omega. \qquad (6)$$

It is the MgB_2 case indeed on the reason that the mass ratio is inversely proportional to the hopping integrals ratio: $M/m \propto t/t_z \propto 10$ (see Ref. 39). For the numerical estimates we will take the experimental value for the optical phonon frequency ω. The energy shift ΔE is negative what means the total lowering of the band minimum of the hole carriers due to the polaron effect. Due to ΔE the energy distance r between the σ- and π-bands is shifted by a frequency dependent contribution which is essential for the electronic sector.

B. Electronic Sector

We start here from the high-temperature paramagnetic phase of MgB_2 system and derive the superconducting critical temperature from the condition of instability of the normal state of correlated σ-electrons with a temperature decrease. Namely, the T_c is governed by the solution of the Bethe-Salpeter equation for a vertex Γ_p in the Cooper channel in the reference frame of the electron pair:

$$\Gamma_p = -T\sum_{n,q}\left[-2t_q + V(p-q)\right]G^{0+}_{\omega_n}(q)\,G^{0-}_{-\omega_n}(-q)\,\Gamma_q, \qquad (7)$$

where $G^{0s}_{\omega_n}(q) = 1/(-i\omega_n+\xi_q)$ is the normal state Green's function for a strongly correlated σ-electron with a spin orientation s and an energy dispersion $\xi_q = ft_q - \mu$ with the correlation factor[40]

$$f = \frac{2w_1+w_2+3r}{5w_1+4w_2}. \qquad (8)$$

The Matzubara frequencies are given by $\omega_n = (2n+1)\pi T$ in Eq. (7).

The expression for the chemical potential

$$\mu = w_1\frac{w_2-5r}{5w_1+4w_2} \qquad (9)$$

follows from the equation $n_\sigma + n_\pi = 2$ for the total electron density per boron at the assumption of a complete ionicity of Mg ions in $Mg^{2+}\left[B^-\left(p^{n_p+n_\sigma}\right)\right]_2$ system. In the integral kernel of Eq. (7) near the Γ-A line of the Brillouin zone the Coulomb vertex $V(p-q)$ can be factorized as

$$V(p-q) = 2\beta t_p t_q, \qquad (10)$$

where the parameter $\beta = V/6t^2$ labels an effect of the Coulomb repulsion for the nearest σ-electrons and the energy dispersion is $t_p = 3t\left[1-\left(p_x^2+p_y^2\right)/12\right]$ near the Γ-A line. The kernel of the integral equation (7) does not contain another kinematic vertices[40] which could be essential for superconducting condensates at moderate densities of carriers and/or for specific symmetries of superconducting order parameters.

Summation over the Matzubara frequencies being performed, Eq. (7) takes the form:

$$1 = \sum_q t_q\frac{1-\beta t_q}{\xi_q}\tanh\frac{\xi_q}{2T_c}. \qquad (11)$$

The superconducting coupling constant can be written as

$$\lambda = \frac{\mu}{w_1 f^2}\left(1-\frac{\beta}{f}\mu\right), \qquad (12)$$

so the chemical potential is restricted by the inequality $0 \le \mu \le f/\beta$ which generates constraints for the electron structure parameters $w_{1,2}$ and r. The larger is the electron-electron Coulomb repulsion $V \sim \beta$ the more narrow becomes the superconducting region and the lower is T_c.

Hereafter we will neglect the inter-electron Coulomb repulsion ($\beta = 0$). Then the superconducting critical temperature satisfies the following equation:

$$1 = \int_{\xi(-w_1)}^{\xi(w_1)} d\xi\ \rho(\xi)\frac{\xi+\mu}{\xi f^2}\tanh\frac{\xi}{2T_c}, \qquad (13)$$

where the $\xi(w_1)$ and $\xi(-w_1)$ are the energy dispersion values of σ-electrons at the top and the bottom of their

energy band, respectively, and $\rho(\xi)$ is the electronic density of states. Eq. (13) for the superconducting critical temperature coincide with one derived in Refs. 31,32 in a different way.

Note that the kinematic superconducting mechanism was also applied to MgB_2 in Ref. 41 but with a non-physical negligence of the π-electrons role. In our approach the great importance has the characteristic energy difference r_0 between of σ- and π-bands, shifted by ΔE due to the interaction between the light holes in the σ-band and the quasi-$2D$ E_{2g} phonons:

$$r = r_0 + \Delta E. \tag{14}$$

Due to the chemical bonds nature the hydrostatic pressure decreases the inter-plane distance more readily than the in-plane boron-boron distance, so that $dw_2/dp \gg dw_1/dp$ (c.f. Ref. 42) and the latter derivative can be neglected in our estimates. The calculations of the $T_c(r)$ demonstrate[31] that $r \gtrsim -w_1/4$ for MgB_2. As the next step we differentiate the integral equation (13) with respect to the pressure and take into account Eq. (14). Assuming the rectangular density of states $\rho = \theta(w_1^2 - \xi^2)/2w_1$ we obtain after the subsequent integration the following expression:

$$\frac{d\ln T_c}{dp} = \frac{1}{5w_1 + 4w_2}\left[\frac{2w_1}{T_c} - 1 - 5\ln\left(\frac{\gamma_0 w_1}{\pi T_c}\right)\right]\frac{dr}{dp}, \tag{15}$$

where $\gamma_0 = \ln C$ with $C = 0.577$ being the Euler's constant. Under the natural assumption $T_c \ll w_1$ one can keep only the first term in the brackets in Eq. (15). Then we arrive at the result:

$$\frac{dT_c}{dp} = \frac{2w_1}{5w_1 + 4w_2}\frac{d\Delta E}{dp}. \tag{16}$$

Taking into account that the Fröhlich coupling constant $\alpha \propto 1/\sqrt{\omega}$ we find then from Eq. (6):

$$\frac{d\Delta E}{dp} = \Delta E\frac{d\ln(\sqrt{\omega}/\gamma)}{dp}. \tag{17}$$

The parameter $\gamma = c/2a$ evidently decreases with the pressure. As to the quasi-$2D$ frequency ω we use the estimate of a quasi-harmonic phonon via the mode Grüneisen parameter $d\ln\omega/dp = G/B_0$. The quantity B_0 is said to be the bulk modulus. It follows then that both ω and $\sqrt{\omega}/\gamma$ increase with the pressure. These findings are compatible with an experimental study[28,43] of Raman spectra and lattice parameters of MgB_2 under pressure at room temperature and with theoretical estimations[44] of the strong influence of the pressure on E_{2g} mode. Finally we conclude that $d\Delta E/dp < 0$ and $dT_c/dp < 0$.

In an explicit form the baric derivative for the superconducting critical temperature can be written as follows:

$$\frac{dT_c}{dp} = -\frac{|\Delta E|}{5 + 4w_2/w_1}\left(-\frac{d\ln\gamma^2}{dp} + \frac{G}{B_0}\right). \tag{18}$$

III. NUMERICAL RESULTS AND CONCLUSIONS

For numerical calculations we estimate the polaron anisotropy parameter as $\gamma = c/2a = 0.57$. The known value of its derivative[36] is $d\ln\gamma^2/dp = 2d\ln(c/2a)/dp \simeq -2.4\cdot 10^{-3}\ GPa^{-1}$. The bulk modulus is measured[45] to be $B \simeq 114\ GPa$. The Grüneisen parameter reported in Ref. 28 equals $G = 2.9 \pm 0.3$ for the measured Raman active E_{2g}-phonon mode with the energy $\hbar\omega = 76.7\ meV$ which we will use in Eq. (18). In particular, the polaron self-energy $\Delta E = -211.4\alpha\ meV$. The realistic value for the MgB_2 energy bandwidths ratio of π- and σ-electrons is $w_2/w_1 \simeq 18/9 = 2$ (see Ref. 46).

Putting these magnitudes in Eq. (18) we obtain the estimate for the derivative of T_c with respect to pressure:

$$\frac{dT_c}{dp} \simeq -\alpha(5.2 \pm 0.9)\frac{K}{GPa}. \tag{19}$$

The uncertainty of the result comes from the experimental deviation errors in the Grüneisen parameter. The comparison with the low pressure result[1,2] $dT_c/dp = -1.2\ K/GPa$ leads at the interval $\alpha = 0.20 - 0.28$ for the Fröhlich electron-phonon coupling constant. The comparison with the result[1,3,4] $-2\ K/GPa$ leads subsequently at the estimate $\alpha = 0.33 - 0.47$.

These results are shown in Fig. 1. The solid line presents the dependence of the baric derivative dT_c/dp on the electron-phonon coupling constant α. The thin solid lines set the experimentally measured values for dT_c/dp and the dashed lines show error bars for α at a taken value of the baric derivative.

We have to note that the numerical results can be changed if one scales the frequency shift of the in-plane mode with the variation of the interatomic bond distance or lattice parameter[47]. Then the Grüneisen parameter takes even the larger value[28] $G = 3.9 \pm 0.4$ and our Eq. (19) takes the form:

$$\frac{dT_c}{dp} \simeq -\alpha(6.9 \pm 1.1)\frac{K}{GPa}. \tag{20}$$

Subsequently, the estimates for the electron-phonon coupling constant will be different as well: $\alpha = 0.15 - 0.21$ for $dT_c/dp = -1.2\ K/GPa$ and $\alpha = 0.25 - 0.34$ for $dT_c/dp = -2\ K/GPa$.

Combining the numerical results for α we may conclude that the electron-phonon coupling constant in MgB_2 is in the range $\alpha = 0.15 - 0.45$ which seems to be quite reasonable interval for the assumed weak-coupling regime of a large hole polaron.

To resume, we proposed a model in which large anisotropic polarons play an important role decreasing the energy distance r between the σ- and π-bands. The polaron anisotropy is governed by the introduced parameter γ which depends on the geometry of the MgB_2 system influenced by the pressure. Besides, the phonon frequency increases with the pressure which results also in a

decrease of the polaron radius. Thus, the superconducting properties of the MgB_2 system are influenced both by the geometry of the crystal and by the polaron well depth and size.

The superconducting instability is driven by the non-phonon kinematic mechanism with T_c depending on the energy difference r between the σ- and π-bands. The quantity r incorporates all electron-phonon effects in our model. As the result the baric derivative dT_c/dp is calculated as a function of the Fröhlich coupling constant α. At realistic values of α our calculations agree with the experiments. It follows from the arguments discussed in the present paper that the pressure measurements provide us with valuable tests to establish the acceptable model for MgB_2, to understand the mechanism of the superconductivity in this material and to estimate its Fröhlich electron-phonon coupling constant.

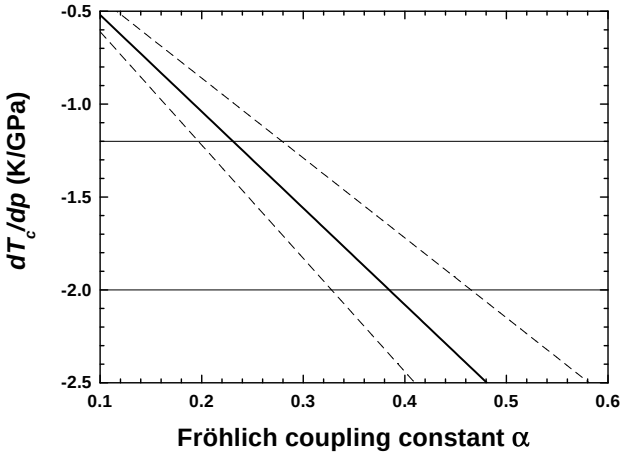

FIG. 1. Dependence of the baric derivative of the superconducting critical temperature on the Fröhlich electron-phonon coupling constant (solid line). The dashed lines show error bars for α due to the uncertainty in the Grüneisen parameter[28]. The thin solid lines show experimental values[1–4] for dT_c/dp.

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